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### FINAL REPORT

on

Merged Beam Studies into the Mechanisms of Hydrogen Molecular Ion Recombination

Prepared for the

U.S. Air Force Office of Scientific Research

by

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## INTRODUCTION.

The aim of this project has been to study the details of the dissociative recombination of hydrogenic molecular ions. These processes serve as important sinks for low energy electrons and sources of excited atoms and molecules in negative ion sources. There has been considerable controversy regarding the rate for the recombination of  $H_3^+$  (v=0) in recent years and this has been a principal concern in this study. The cross section for this process was measured during a previous grant period and it was found that indeed the rate for v=0 ions is about an order of magnitude lower than previous measurements had indicated. This finding is in agreement with the published experiment of Adams et al (1983). It is however in disagreement with the revised estimate  $^{(2)}$  of  $1 \times 10^{-10}$  cm  $^3$  sec  $^{-1}$  published by Adams and Smith in 1988, following a recalibration of their apparatus. We have made a number of new measurements which are pertinent to this topic and these are discussed here.

During this grant period, we have brought a new merged beam apparatus, MEIBE II on-line and have used it to remeasure the  $H_3^+$  recombination, to make a more detailed study<sup>(3)</sup> of the dissociative excitation of  $H_3^+$  and to perform a high energy resolution measurement<sup>(4)</sup> of the dissociative recombination of  $H_2^+$  (v=0). We have also performed the first experimental measurement<sup>(5)</sup> of the cross sections for the dissociative recombination and excitation of  $HeH_3^+$ .

In an experiment performed a few years ago, the branching ratio for the dissociative recombination of  $H_3^+$  in a variety of vibrational states was determined<sup>(6)</sup> using a technique which

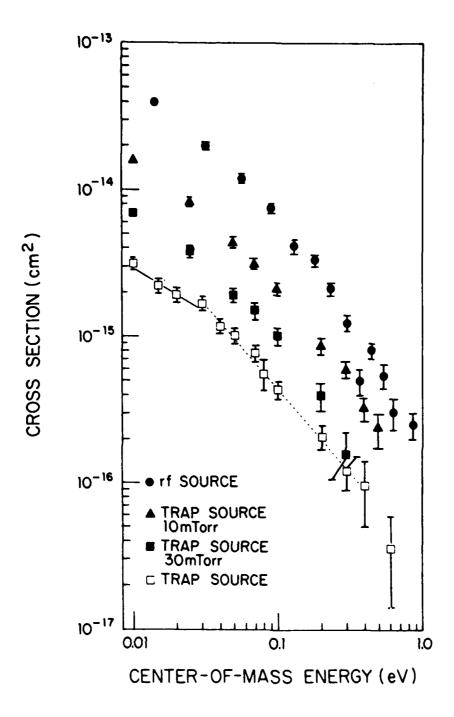
involves placing a grid with known transmission in front of the detector to differentially restrict access to the detector by differing numbers of particle sets. This experiment has been repeated  $^{(7)}$  for ground vibrational state  $\mathrm{H}_3^+$  ions and the results are discussed here.

One of the goals of this experimental program is to identify the excitation state of the recombination products and to this end, a time and position sensitive detector is currently under testing in the laboratory. The operation of this detector and progress to date will be discussed.

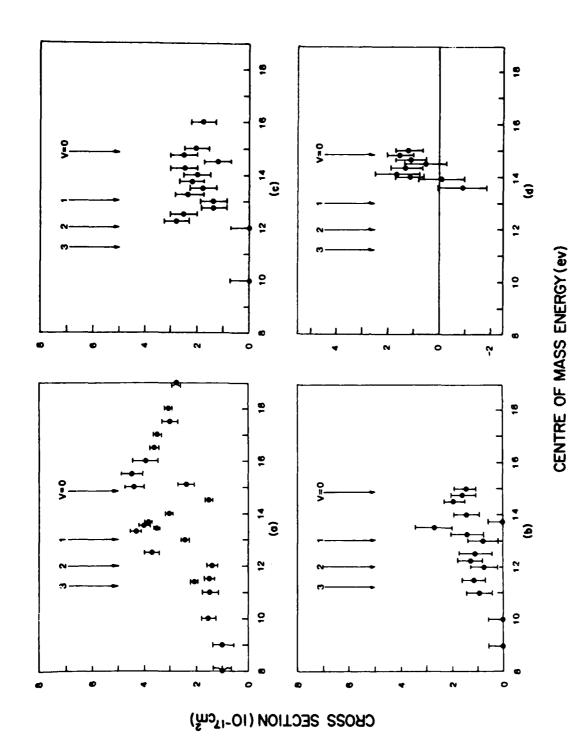
Finally, although sponsored under a separate AFOSR grant, mention should also be made of the International Symposium on Dissociative Recombination: Theory, Experiment and Applications which was held at Lake Louise, Alberta in May, 1988 as it bears direct relevance to this project. The proceedings of this symposium have been published as a book. (8)

# I. THE DISSOCIATIVE RECOMBINATION OF $H_3^+(v=0)$ .

During the last contract period, cross sections for the dissociative recombination of  $\mathrm{H}_3^+$  ions with varying vibrational state distributions were measured<sup>(9)</sup> using the merged beam technique and the results are displayed in figure 1. Control of the vibrational populations was made possible by varying the operating parameters of the r.f. trap ion source used to produce the ions. The internal energies of the ions were determined by studying the observed threshold energy for the dissociative excitation of the ions by electron impact, fi ure 2. It can be seen that for ions with the lowest attainable internal energy, the measured cross sections were about an order of magnitude smaller



 Cross sections for e-H<sub>3</sub><sup>+</sup> recombination. The four curves shown here were measured using ions formed under conditions which yielded the corresponding four excitation functions shown in fig. 2. (From ref.9).



 $e+H_3^+ + H_3^+ (^3E^-) + e + H_2^+ + H^+ + e$ . (a) Ions produced in a conventional rf source at 100 mtorr. (b) Ions produced in trap source at 10 mtorr; pure  $H_2$ . (c) Trap source 30 mtorr; pure  $H_2$ . Trap source, 70 mtorr, sections for the dissociative excitation low extraction 10:3:1,  $H_2:$  Ar: He mixture. (From ref.9) Cross 7

than those for ions produced using a conventional radio frequency ion source with a range of vibrational states in excess of v=4 populated. The rate coefficient, calculated from these cross sections has a value of about  $1 \times 10^{-8} \, \mathrm{cm}^3 \, \mathrm{sec}^{-1}$  at 300K in agreement with the original published values of Adams et al<sup>(2)</sup>, measured using the Flowing Afterglow Langmuir Probe, (FALP), technique. Adams and Smith<sup>(3)</sup> have however recalibrated their apparatus and now claim that the rate coefficient for the recombination of  $\mathrm{H}_3^+$  (v = 0) is of the order of  $1 \times 10^{-10} \, \mathrm{cm}^3 \, \mathrm{sec}^{-1}$  at room temperature.

This recalibration involved observing the recombination of the ions He and HeH and comparing these with that for H<sub>2</sub>. It was found that in fact the rate of electron decay measured in the FALP apparatus was the same for all three ions suggesting that they had similar recombination rates. Since He is an atomic ion, it can only undergo radiative decay at low temperatures and the rate calculated (10) for <10<sup>-11</sup>cm<sup>3</sup>sec<sup>-1</sup>. Although HeH<sup>+</sup> is molecular in calculations (11) have shown that there is no available curve crossing for this ion through which direct dissociative recombination can proceed. It has always been assumed therefore that it should also have a negligible recombination rate. From their observations, Smith and Adams (12) therefore proposed that  $H_2^+$  (v = 0) also would have a recombination rate coefficient of the order of 10<sup>-11</sup>cm<sup>3</sup>sec<sup>-1</sup> although they have since revised<sup>(3)</sup> this figure to less than  $1 \times 10^{-10} \text{cm}^3 \text{sec}^{-1}$  since the sensitivity of their technique is insufficient to define the value to such precision.

The publication of this revised estimate for the  $H_3^+$  rate has prompted us to look at this problem in more detail and we have

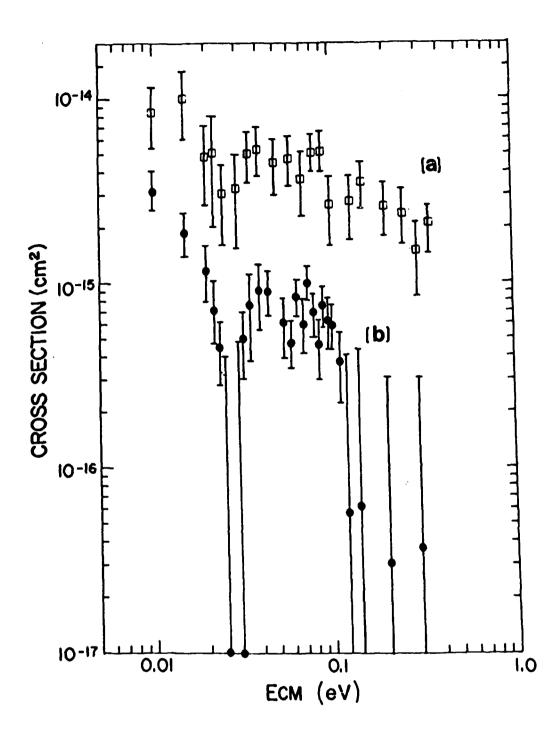
performed measurements on the dissociative recombination and excitation of  $HeH^+$ , the dissociative excitation of  $H_3^+$  and the branching ratio for the recombination of  $H_3^+$  in order to shed some light on the process.

1. Measurements of the Dissociative Recombination and Excitation of HeH<sup>+</sup>.

As mentioned above, the HeH<sup>+</sup> recombination has always been assumed to be negligible but in fact no experimental measurements of the rate had been performed. We have corrected this deficiency and the results for cross section measurements as a function of energy are shown in figure 3. The upper data set refers to ions which were formed under high ion source extraction conditions while the lower set refer to low extraction ions. (The ion source has an extraction electrode the potential of which with respect to the source body can be varied. Experience has shown that ions extracted under high, (100V), potential conditions tend to be excited while a low potential, (10V), on this electrode leads to de-excited ions).

It can be seen that neither sets of cross sections are negligible. In fact the lower data correspond to a rate coefficient of between  $5 \times 10^{-9}$  and  $1 \times 10^{-8} \text{cm}^3 \text{sec}^{-1}$ , the exact value depending upon assumptions concerning the form of the cross section above 0.1eV. In order to determine the internal energies of the ions, the electron impact dissociative process:-

$$e + HeH^{+} \rightarrow H + He^{+} + e$$



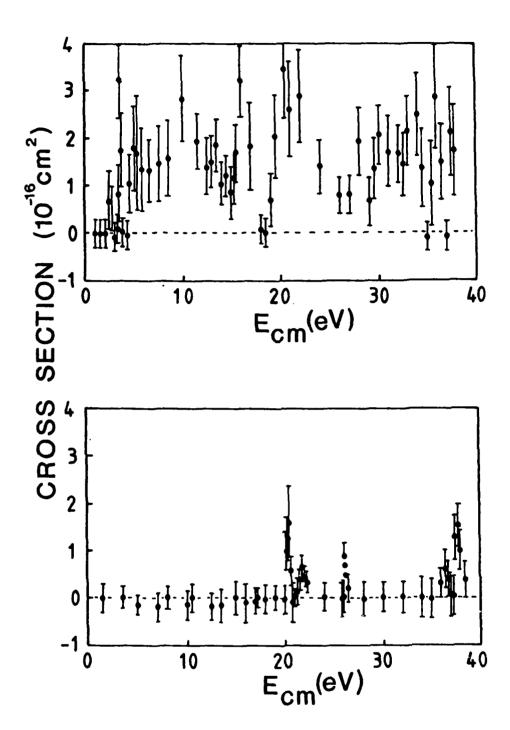
 Measured dissociative recombination cross sections for HeH<sup>+</sup> ions prepared under (a) high extraction and (b) low extraction conditions. (from ref.5). was examined. The results of these measurements are shown in figure 4. The upper data display a threshold in the vicinity of 3eV indicating that the ions are in an electronically excited state, (see figure 5). The lower set however display thresholds at 20, 26 and 36eV corresponding to transitions from the v=0 level of the ground electronic state of HeH<sup>+</sup> to the first three excited states, (see figure 5). Thus it can be said that the cross section for the recombination of HeH<sup>+</sup> is not in fact negligible and this indicates that the recombination must proceed via an alternative mechanism such as rydberg state capture, (indirect recombination). It is our intention to study this further at a later date.

It should be mentioned that attempts to measure a cross section for the recombination of He<sup>+</sup> ions were unsuccessful as expected given that the calculated rate for this process is so very small, thus confirming the efficacy of our measurement technique.

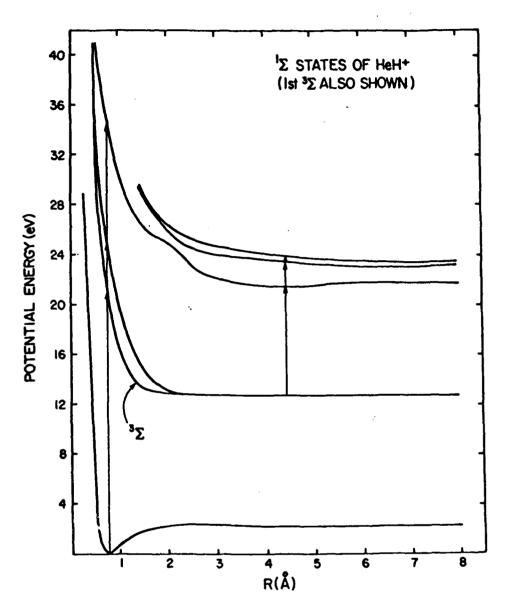
This work has been written up and accepted for publication in Physical Review A.

# 2. The Dissociative Excitation of $H_3^+$ .

In our published measurement of the dissociative recombination of  $H_3^+$  shown in figure 1, there was some ambiguity concerning the vibrational state population for the lowest internal energy ions. This arose because the observed threshold for the excitation process, figure 2d, occurred below the energy expected for a transition from the v=0 level of the ground state to the upper  $^3E'$  state, (see figure 6). At the time it was



4. Measured dissociative excitation cross sections for HeH<sup>+</sup> ions prepared under (a) high extraction and (b) low extraction conditions. (from ref.5).



5. Potential energy curves for the ground and excited  $^{1}\Sigma$  states of HeH $^{+}$  and the first  $^{3}\Sigma$  state $^{(18)}$ . The arrows indicate the transitions seen in the electron impact excitation results. (From ref.5).

believed that the sub-threshold cross section might have arisen from transitions from ions in the v=1 level and so it was stated that the recombination was performed using ions with v=0 and v=1 populated. Repeated measurements of the recombination cross section using the new MEIBE II apparatus confirmed that indeed the cross sections shown in figure 1d were the lowest attainable. A decision was made to re-examine the dissociative excitation process in more detail and the results of this measurement are shown in figure 7.

It is immediately obvious that there is considerable resonant structure in this cross section and this must be due to a process akin to dielectronic recombination in which the incoming electron, with energy just less than that required to excite a given ion state, is resonantly captured into a neutral rydberg state lying just below the ion state. In this case however, the rydberg state is predissociated by the repulsive ion state and so the recombination is not stabilized. Similar resonant structures have been seen by us in the dissociative excitation of  $N_2^+$  (13) and HeH<sup>+</sup>, (fig 4). In the latter case in particular, the process is dominated by the resonances, direct excitation being negligible in the threshold region. This is similar to the situation for the ionization of molecular hydrogen which is dominated by resonant autoionization at threshold. Interpretation of the role of resonances in the dissociative excitation process will require theoretical analysis and it is to be hoped that some theoretician will tackle this problem in the near future.

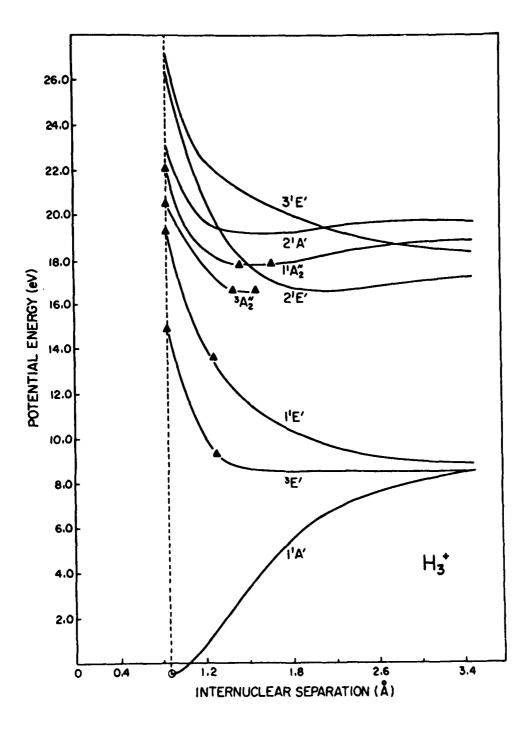
Of particular significance in the present context is the fact that the structure appearing at 14eV, lying just below the threshold for excitation of the  $^3E'$  state from the v=0 level of the ground state, had previously been interpreted by as us as evidence for the presence of v=1 states in our beam. Clearly this resonance is too narrow and at the wrong energy to be due to excitation from the v=1 level. This means therefore that the recombination results shown in fig. 1 must refer to ions in the ground vibrational state only.

# 3. Branching Ratio for $H_3^+$ (v = 0) recombination.

In a previous measurement  $^{(6)}$ , the branching ratio for the dissociative recombination of  $\mathrm{H}_3^+$  ions in a variety of vibrational states was determined using a technique which involves inserting a grid with known transparency, t in front of the neutral detector. Since the probability of n particles traversing the grid is proportional to  $\mathrm{t}^n$ , different decay channels producing different numbers of particles can be distinguished from each other. Normally the dissociation products arrive simultaneously at the detector and appear as a single particle carrying the full beam energy, (usually 400 KeV). The presence of the grid means that often, not all of the particles will reach the detector and so signal peaks will be detected at fractional energies,  $\mathrm{E/n}$ ,  $\mathrm{2E/n}$  etc. For the case of the three possible decay channels for  $\mathrm{H}_3^+$  namely:

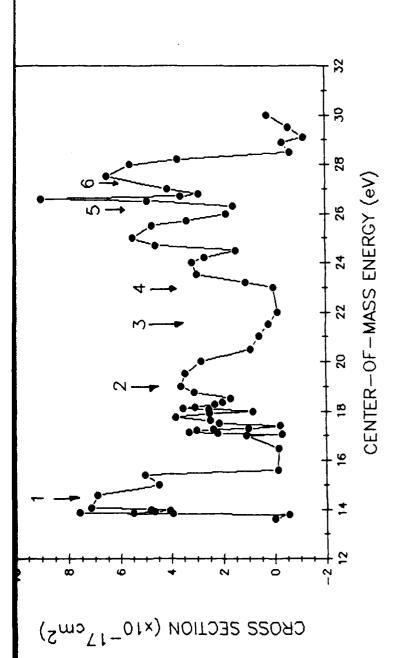
$$e + H_3^+ \rightarrow H + H + H$$
 I
$$\rightarrow H_2 + H$$
 II
$$\rightarrow H_3^*$$
 III

producing  $N_{I}$ ,  $N_{II}$  and  $N_{III}$  counts per second respectively, the count rate registered as 1/3, 2/3 and full beam energy pulses will be:

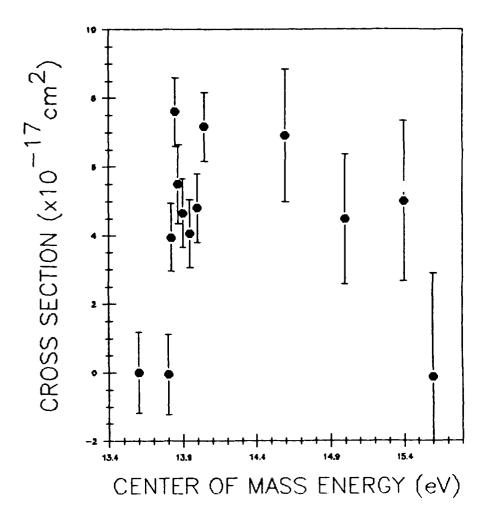


6. Potential energy curves for H<sub>3</sub><sup>+</sup> in D<sub>3h</sub> symmetry.

Singlet states (ref.19), Triplet states (ref.20), Triangles (ref 21).



electron impact. Excitation thresholds for the states (1)  $^3\!\mathrm{E}',$ (2)  $^{1}E'$ , (3)  $^{1}A_{2}$ , (4)  $^{2}A_{1}$ , (5)  $^{2}E'$ , (6)  $^{3}E'$  indicated by 7a. Cross sections for the dissociative excitation of  $\mathrm{H}_3^+$  ions by arrows. (From ref. 3).



7b. Enlarged view of the threshold region of 7a with error bars shown.

$$n_1 = 3t(1-t)^2N_1 + t(1-t)N_{II}$$
 $n_2 = 3t^2(1-t)N_1 + t(1-t)N_{II}$ 
 $n_3 = t^3N_1 + t^2N_{II} + tN_{III}$ 

In the previous measurement, channel III was neglected as it was assumed to be negligible. In this experiment it has been included and in fact turns out to be significant.

Measurements have been performed  $^{(7)}$  for one energy only,  $(E_{\rm cm} = 0.01 {\rm eV})$  and the following cross sections for the individual channels have been measured:-

$$\sigma_{\text{I}}$$
 = 1.85 x 10<sup>-15</sup> cm<sup>2</sup> ± 30%  
 $\sigma_{\text{II}}$  = 1.44 x 10<sup>-15</sup> cm<sup>2</sup> ± 25%  
 $\sigma_{\text{III}}$  = 0.295 x 10<sup>-15</sup> cm<sup>2</sup> ± 47%  
 $\sigma_{\text{Tot}}$  = 3.6 x 10<sup>-15</sup> cm<sup>2</sup>

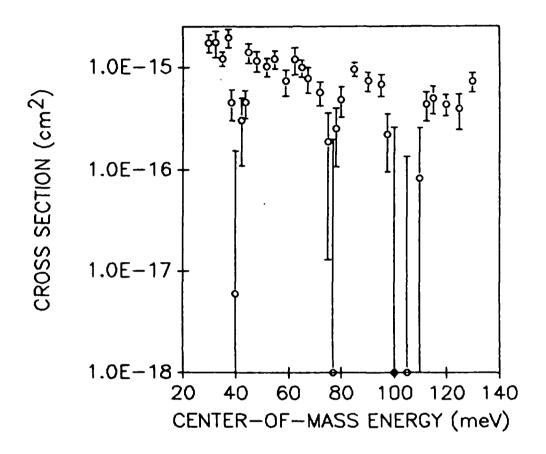
The observation that the channel producing  $H_3^{\star}$  should account for 8% of the total cross section is startling. If direct recombination is ruled out for  $H_3^{\dagger}$  (v=0), then the only explanation for the cross sections shown in fig.1 is "indirect recombination", i.e. recombination occurring via capture into vibrationally excited rydberg states. Normally one would expect these states to be very short lived unless a stabilization mechanism is available. It is not clear at the present time what this mechanism might be but this measurement clearly shows that it is somehow possible for

these states to relax to lower, non-autoionizing rydberg states which are sufficiently long-lived to reach the detector. The flight time for this to occur in the MEIBE apparatus is approximately  $10^{-7} {\rm sec.}$  (It is possible that these high lying states have sufficient overlap with the repulsive ground state of  ${\rm H_3}$  to be predissociated by that state leading to H + H + H or H<sub>2</sub> + H products). This would explain the substantial recombination cross section measured in our experiments. In the high pressure environment of the FALP apparatus however, these  ${\rm H_3^*}$  molecules may be rapidly reionized and so a negligible recombination cross section would be measured. So far these are just speculations and much more experimental and theoretical work is called for before this problem is fully understood. We do however appear to be on the right track.

# II. THE DISSOCIATIVE RECOMBINATION OF $H_2^+$ .

During the last grant period, we published cross sections for the dissociative recombination of  $\mathrm{H}_2^+$  ions in low vibrational states  $^{(14)}$  and found sharp resonances due to rydberg state interactions. They had been predicted theoretically, (see discussions in ref. 8). The energy separations of the data points in this measurement were rather large so it was decided to repeat the measurement using a much finer separation. It was also decided to perform this measurement with the new MEIBE II apparatus to observe its energy resolution. The results of this experiment are displayed in figure 8. Three sharp window resonances are observed between 20 meV and 100meV and the width of the narrowest of these is observed to be of the order of 5meV! This is really excellent

# DISSOCIATIVE RECOMBINATION OF H2+



8. Cross sections for the dissocciative recombination of H<sub>2</sub><sup>+</sup> (v=0) measured using the MEIBE II apparatus. The resonance at 40meV is seen to be ~5 meV wide illustrating the high resolution capabilities of the merged beam technique. (From ref.4).

energy resolution and clearly demonstrates the power of the merged beam technique. The absolute position of these resonances has yet to be verified since the energy calibration of the MEIBE II apparatus has not yet been performed. This will be done shortly and at that time these results will be submitted for publication (4).

### III. FINAL PRODUCT STATE DETERMINATION STUDIES.

The dissociative recombination of  $H_2^+$  leads to the formation of hydrogen atoms in a variety of excited states thus:-

$$e + H_2^+(v) \rightarrow H(1s) + H(nl)$$

Very little is known about the probability for forming a given final state, the only experimental studies being those of Vogler and Dunn <sup>(15)</sup> and Phaneuf et al <sup>(16)</sup> who measured partial cross sections for the formation of D(2p) and D(n=4) atoms respectively. These measurements however were performed on ions with a wide range of vibrational states populated and for electron energies in excess of 0.5eV. It is our intention to examine this problem using ions with selected vibrational state populations and at low electron energies.

The approach being adopted is to measure the kinetic energy of the products released in the recombination using a time and position sensitive detector and hence to determine the potential energy of the atoms. For electrons with energy  $\mathbf{E}_{\mathrm{Cm}}$  colliding with  $\mathbf{H}_2^+$  ions with vibrational energy  $\mathbf{E}_{\mathbf{V}}$  above the ground vibrational state, the kinetic energy released to the products is:-

$$E_{cm} + E_{v} + \Delta [E(H_{2}^{+}, v=0) - E(H(1s) + H(n1))]$$

For low energy electrons and ground state ions, only the H(1s) + H(2l) channel is open since there is no efficient mechanism leading to H(1s) + H(1s). As the vibrational energy of the ions and/or the electron energy is raised, other channels open up. The energy released for specific channels from specified vibrational levels is tabulated in Table 1.

The principle behind the technique of determining the internal energies of dissociation products has been explained fully by de Bruijn and Los<sup>(17)</sup> and it is not necessary to repeat this here. The detector that has been constructed is illustrated in figure (9) and the accompanying electronic circuitry in fig. 10. This has undergone extensive electronic testing and first measurements of coincident particle detection will be performed very shortly.

The measured time and position resolutions attainable with this device are ~0.5ns and  $100\mu m$  respectively, corresponding to an energy resolution of 0.05eV. This will allow the products from individual vibrational states of the molecular ion to be identified.

## FURTHER STUDIES.

It is our intention to continue these studies and in particular to perform further high resolution studies of  $H_2^+$  and  $HeH^+$ , measurements of the branching ratio for the recombination channels of  $H_3^+$  and its isopotomers,  $H_2D^+$ ,  $HD_2^+$  and  $D_3^+$ , and of the excitation of molecular ions. Measurements of the excitation states of the products of diatomic recombination are planned for the near future. Details of these projects are to be found in ref 22.

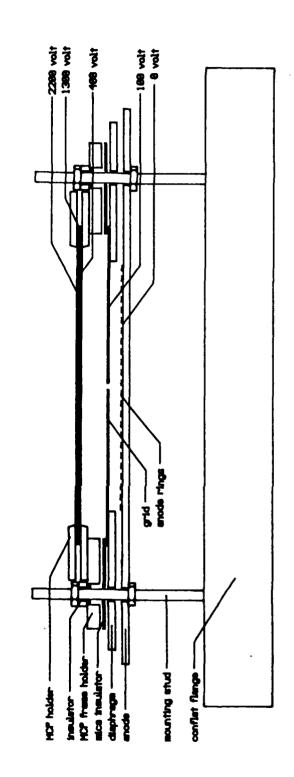
TABLE 1. Final channel kinetic energy release for e +  $H_2^+$  (v) recombination at  $E_{\rm cm}$  = 1.0 eV.

K.E. RELEASED (eV)
Final Channel

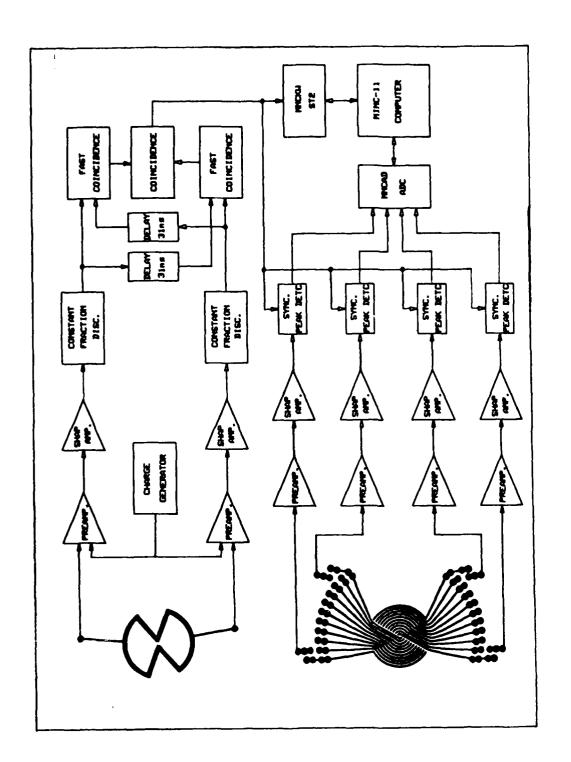
	v=0	v=1	v=2	v=3	v=4	v=5	v=6
H(1s) + H(21)	1.75	2.02	2.26	2.51	2.72	2.96	3.13
H(1s) + H(31)	-	0.13	0.37	0.625	0.835	1.08	1.25
H(1s) + H(41)	-	-	-	-	0.17	0.41	0.58
H(1s) + H(51)	_	_	-	-	-	0.11	0.28

More channels open up as  $\mathbf{E}_{\mathrm{cm}}$  is increased.

# Position and Time Sensitive Detector



kinetic energy of the products of diatomic molecule recombination. Time and position sensitive detector for the measurement of the (MCP -Multi-Channel Plate detector). ъ.



10. Circuit diagram for the operation of the time and position sensitive detector.

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- 3. Merged Beam Studies of Dissociative Recombination Recent Results J.B.A. Mitchell and F.B. Yousif in 1. p109.
- 4. Laser Stimulated Radiative Recombination: A Field ionization Approach. T.J. Morgan and J.B.A. Mitchell in 1. p175.
- 5. Molecular Ion Recombination: Branching Ratio Measurements.
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- 11. High Resolution Studies of  $H_2^+$  Dissociative Recombination.

  P. Van der Donk, F.B. Yousif and J.B.A. Mitchell

  (In preparation).